

Thermal Gelation Properties of Methyl and Hydroxypropyl Methylcellulose*

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Synopsis

Aqueous solutions of methyl and hydroxypropyl methylcellulose are known to gel upon heating. These gels are completely reversible in that they are formed upon heating yet will liquefy upon cooling. The precipitation temperature, gelation temperature, and gel strength of these methylcellulose solutions were determined as a function of molecular weight, degree of methyl and hydroxypropyl substitution, concentration, and presence of additives. The precipitation temperature of these polymer solutions decreases initially with increasing concentration until a critical concentration is reached above which the precipitation temperature is little affected by concentration changes. The incipient gelation temperature decreases linearly with concentration. The strength of these gels is time dependent, increases with increasing molecular weight, decreases with increasing hydroxypropyl substitution, and depends on the nature of additives. Hydrophobe-hydrophobe interaction or micellar interaction is postulated to be the cause of gelation. This thermal gelation property of the polymers is utilized in many end uses including food, pharmaceuticals, ceramics, tobacco, and other industrial applications.

INTRODUCTION

Thermally reversible gelation of aqueous solutions of macromolecules has been characterized as due to the formation of a three-dimensional crosslinked network structure.¹ Since this sol-gel transformation is reversible within a narrow temperature range, it does not involve the making or breaking of any covalent bonds, and the quasi-crosslinkages in the gel network structure are due to secondary valence forces as the solvent power of the medium decreases. Most macromolecules in solution exist as randomly coiled isolated chains. When the temperature is decreased or increased beyond the gelation temperature, and at appreciable concentrations, the polymer begins to reconstitute the original solid-state structure. Gelation is therefore an intermediate nonequilibrium metastable state in which a three-dimensional network structure is formed due to secondary valence forces. The mobility of the chain is greatly restricted and the reconstitution of the original solid state becomes an extremely slow process.

Some of the classical examples of natural polymers exhibiting the sol-gel transformation phenomenon are gelatin (collagen protein) and carrageenan (acidic polysaccharide). In these cases polymers exist as three-stranded (collagen) or two-stranded (carrageenan) helical rods which go into solution as random coils at elevated temperature. Upon cooling, a continuous network is formed due to partial formation of the helix.^{2,3} Several synthetic polymers are also known to gel either in aqueous or organic medium. In many cases gels ex-

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hibit syneresis as the polymer network contracts on standing, liberating the pure solvent.

In contrast to these polymers, aqueous solutions of methylcellulose are known to gel when the temperature is increased.⁴ Gelation of methyl or hydroxypropyl methylcellulose solution is primarily caused by the hydrophobic interaction between molecules containing methoxyl substitution. In a solution state at lower temperatures, molecules are hydrated and there is little polymer-polymer interaction other than simple entanglement. As the temperature is increased, the molecules gradually lose their water of hydration, which is reflected by a drop in the relative viscosity. Eventually, when a sufficient but not complete dehydration of the polymer occurs, a polymer-polymer association takes place and the system approaches an infinite network structure reflected by a sharp rise in relative viscosity. These gels are completely reversible in that they are formed upon heating yet will liquefy to the original consistency upon cooling.

There exists a certain degree of controversy regarding the mechanism of gelation of methylcellulose. The possibility of the gelation being caused by the presence of "long unaltered residues of original cellulose structure," as suggested by Ott and Spurlin,⁵ is unlikely because for highly substituted methylcellulose, such as the one studied by Heyman,⁴ there can be relatively few unsubstituted monomer units present. Solubility of methylcellulose, on the other hand,⁵ is highly dependent on the uniformity of substitution. For example, uniformly substituted methylcelluloses prepared homogeneously from sodium cupricellulose or in quaternary base are water soluble at a much lower degree of substitution than are the ones made from alkali cellulose.

Observing that the gelation temperature increases with the decrease in methyl substitution, Vacher⁶ concluded that the lack of trisubstitution was the cause of gelation. A lower gelation temperature can also be interpreted as resulting from increased hydrophobe-hydrophobe interaction with increasing methyl substitution. Rees⁷ therefore accurately described these gels as "micellar gels" analogous to the nonionic surfactants, which also exhibit cloud points on heating.

The thermal gelation property of methyl and hydroxypropyl methylcellulose has promoted their use in a myriad of applications. Gelled, these cellulose ethers provide the binding and "green strength" necessary for workability for heat-setting reconstituted tobacco sheets and for extrusion of ceramic bodies. In many food applications the gelation properties of these cellulose ethers are used as binders and extrusion aids for reconstituted meat and vegetable products, foam stabilizers for foam mat drying, dough strengtheners for low-gluten bakery products, and grease barriers for fried foods. In many adhesive, coating, and printing techniques, the temporary gelation of these cellulose ethers provides viscosity control for high-temperature curing. In suspension polymerization, the gelation of these adsorbed cellulose ethers on the monomer droplets provides the stability of the suspension. In pharmaceutical application, gelation and gel strength of these ethers find their use in tablet coating and the manufacture of medicinal capsules.⁸ A proper characterization of the gelation properties of these cellulose ethers is therefore essential in order to better utilize these properties in different end use applications.

EXPERIMENTAL

Material

Methylcellulose and hydroxypropyl methylcellulose samples used in this study were either commercial samples (Methocel* brand cellulose ethers) or samples made in the laboratory according to the procedure outlined in the literature.⁹ Samples of methyl and hydroxypropyl methylcellulose were made having various degrees of substitution (D.S.) and molecular weights. Some of the commercial products used in this study are described at the top of Table I. In addition, the relationship between molecular weight and 2% aqueous solution viscosity at 20°C is also shown since it is convenient to describe molecular weight in terms of viscosity of a solution.

Procedures

The effects of shear rate and temperature on the viscosity of the solutions were obtained by using a Haake Rotovisco viscometer Model RV-3 (Haake, Inc., Saddlebrook, N.J.). Temperature and rate of temperature changes in the solutions during viscosity measurements were controlled by using a Tamson circulating bath fitted with a Neslab temperature programmer TP-2 (both from Neslab Instruments, Inc., Portsmouth, N.H.). The incipient gelation temperatures (IGT) of methylcellulose solutions were obtained by measuring the viscosity of the solution at a constant rate of shear as a function of temperature utilizing the temperature programmer at 0.25°C/min. The temperature at which the viscosity reaches a minimum is called the IGT. At or near the IGT, excessive shearing may break up the gel, giving a wrong gel point. As the temperature approaches the IGT, the rotation of the bob is stopped; and at increments of 2°C,

TABLE I
Substitution and Viscosity Ranges of Methocel Brand Cellulose Ethers^a

Product designation suffix	Methoxyl degree of substitution DS	Hydroxypropyl molar substitution MS
A (η)	1.6-1.8	0
F (η)	1.6-1.8	0.1-0.2
E (η)	1.65-1.9	0.2-0.3
K (η)	1.1-1.4	0.1-0.3
η , cP	2% Viscosity range, cP	M_w Range
5	4-6	18,000- 22,000
25	20-30	48,000- 60,000
50	40-60	65,000- 80,000
100	80-120	85,000-100,000
400	350-550	120,000-150,000
1500	1200-1800	170,000-230,000
4000	3500-5500	300,000-500,000

^a η Denotes viscosity of 2% solution at 20°C. The viscosity ranges and the corresponding molecular weight ranges are given in lower part of table.

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the bob is turned on just for a few seconds to obtain the viscosity reading, thus minimizing gel structure breakdown.

Precipitation temperatures of methylcellulose solutions were determined by measuring the light transmission of the solution as a function of temperature using a Brinkman PC/1000 colorimeter (Brinkman Instruments, Westbury, N.Y.) which was modified in the following manner. The apparatus consists of 2.5-in.-diameter and 4.25-in.-long cylindrical aluminum block wound with 0.25-in.-O.D. copper tube through which water is circulated from a constant-temperature bath fitted with a temperature programmer as described before. The block has a 0.5-in.-diameter and 3-in.-long cavity at the center into which methylcellulose solution contained in a 13-mm-O.D. and 11-mm-I.D. Pyrex glass ampoule is placed. The ampoule is sealed to eliminate evaporation loss during heating. Two $\frac{1}{8}$ -in.-diameter flexible fiber glass light guides (Ealing Corp., South Natick, Mass.) are placed one at each end of the ampoule through the aluminum block making sure that the light guide tips are perfectly aligned and are perpendicular to the ampoule. The other ends of the light guides are connected to the colorimeter, one to the light source and the other to the detector. The wavelength of light selected was 545 nm, at which minimum absorbance of light by methylcellulose solution was observed. The temperature of the solution was measured by placing a thermocouple in the aluminum block close to the ampoule. Initially, as the temperature was increased at a rate of 0.25°C/min, the light transmission remained at 100%, and then at some elevated temperature the light transmission began to decrease with increasing temperature. The temperatures at which light transmission reached 97.5% and 50% are called incipient precipitation temperature (IPT) and cloud point (CP), respectively.

Gel strengths of aqueous methylcellulose gels formed at 65°C were measured by using a cone Penetrometer (Precision Scientific Company, Chicago, Ill.) following the procedures of Haighton.¹⁰ Cones were made of aluminum with tooled steel tips. Solutions contained in covered glass dishes were heated in a water bath at 65°C, and then the depths of penetration of the cones were measured by allowing the cone, preheated in a 65°C oven, to penetrate for 300 sec. During the measurement the gel dish and the cone were placed in a specially designed oven maintained at 65°C. The yield value of the gel was calculated from the following equation:

$$\tau_y = Kmg/h^2 \quad (1)$$

where τ_y is the yield value, g/cm²; m is the weight of the cone and the plunger rod; g ; g is acceleration due to gravity; h is depth of penetration, cm; and k is the constant which depends only on the angle of the cone.

An exact expression for the constant K has been derived by Agranant and Volarovich,¹¹ and their equation is based upon a lengthy analysis of the plastic deformation during penetration. The Agranant equation was used to calculate the values of K , as shown in Table II.

TABLE II
Values of Constant K for Cones with Different Angle of the Cone

Cone angle	30°	45°	60°	90°	120°
K	0.959	0.416	0.214	0.0995	0.04

RESULTS AND DISCUSSIONS

Figure 1 is a curve showing the typical relationship between viscosity and temperature for a methylcellulose solution. As the temperature is increased, the viscosity of the solution decreases initially until the temperature reaches the incipient gelation temperature at which there is a sharp rise in viscosity, indicating gelation. The cooling curve, on the other hand, looks quite different. The viscosity increases on cooling, reaches a maximum, and then decreases until a slope inflection is reached as it merges with the original heating curve. Since gelation is a time-dependent kinetic phenomenon, the rates of heating and cooling and the rate of shear are very important in determining the shape of the curve. Ideally, in the absence of any large shearing forces and when the heating and cooling rates are extremely small, the minimum in the heating curve and the maximum in the cooling curve should appear at the same temperature, representing the true gelation temperature. In actual practice, it is very difficult to measure this true gelation temperature. As shown in Figure 1, the minimum in the heating curve and the maximum in the cooling curve could be 8–10°C apart, and the true gelation temperature is somewhere between the two temperatures.

Another phenomenon that is observed for the methylcellulose solutions on increasing temperature is the precipitation of the molecules as observed by the light transmission measurement. Figure 2 shows a typical precipitation phenomenon for aqueous solutions of a hydroxypropyl methylcellulose sample at different concentration. As the temperature is increased, the light transmission remains unchanged until, at the IPT, the light transmission deviates from 100% and sharply decreases on increasing temperature.

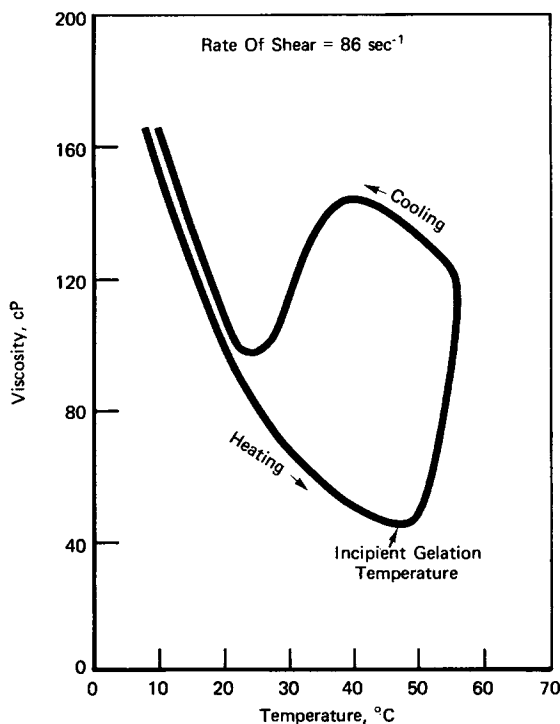


Fig. 1. Gelation of 2% aqueous solution of Methocel A100 on heating at 0.25°C/min.

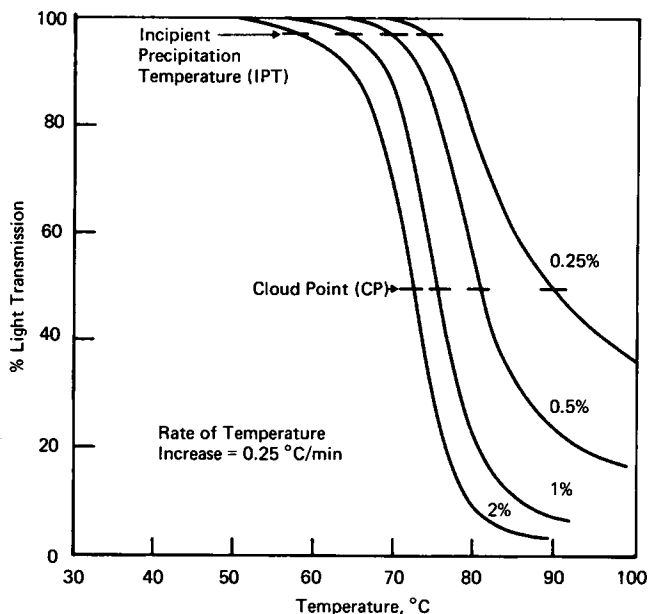


Fig. 2. An illustration of the change of 545 nm light transmission for aqueous solutions of a hydroxypropyl methylcellulose sample having a DS = 1.45 and MS = 0.11, as function of temperature at different concentrations.

This decrease in light transmission actually reflects the initial precipitation of the high molecular weight fractions. As the temperature is further increased, gradual fractionation of the molecules takes place. From that point of view neither the incipient precipitation temperature nor the cloud point actually reflects the precipitation temperature of the average molecular weight of the sample, but rather the precipitation temperature of an unknown molecular weight somewhat higher than the average molecular weight.

Figure 3 illustrates that both IPT and CP decrease initially as the concentration is increased; and then, after a certain critical concentration is reached, the precipitation temperatures are little affected by the concentration.

On the other hand, the incipient gelation temperature exhibits a linear relationship with concentration, as shown in Figure 4. It is also observed from Figure 4 that up to a concentration of about 6.5%, the hydroxypropyl methylcellulose solution becomes turbid before gelation occurs, while above 6.5% concentration the solutions gel before a turbidity becomes visible. Thus, it is conceivable that a methylcellulose solution of the right concentration could form a clear gel even at room temperature. This is in fact observed for low molecular weight methylcellulose solutions. This phenomenon is also true for high molecular weight methylcellulose solutions. But because of the physical difficulty in preparing highly concentrated solutions of high molecular weight methylcellulose, this phenomenon is difficult to observe.

As was mentioned earlier, gelation is a time-dependent phenomenon. When a solution of methylcellulose is heated above the incipient gelation temperature, the gel strength develops gradually with time. Figure 5 shows the rate of gel strength development for a 2% solution of methylcellulose at 65°C. It is observed that the gel strength increases with time and reaches the maximum gel strength

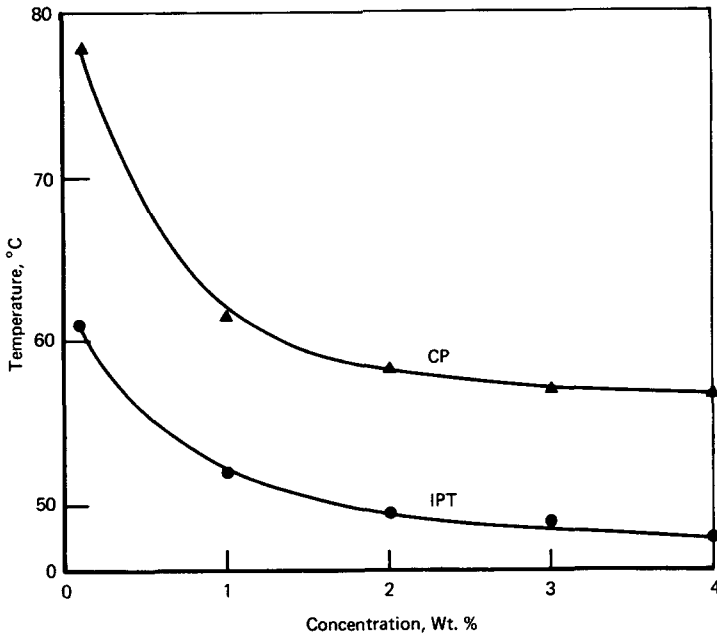


Fig. 3. Incipient precipitation temperature (IPT) and cloud point (CP) of Methocel F50 as function of concentration.

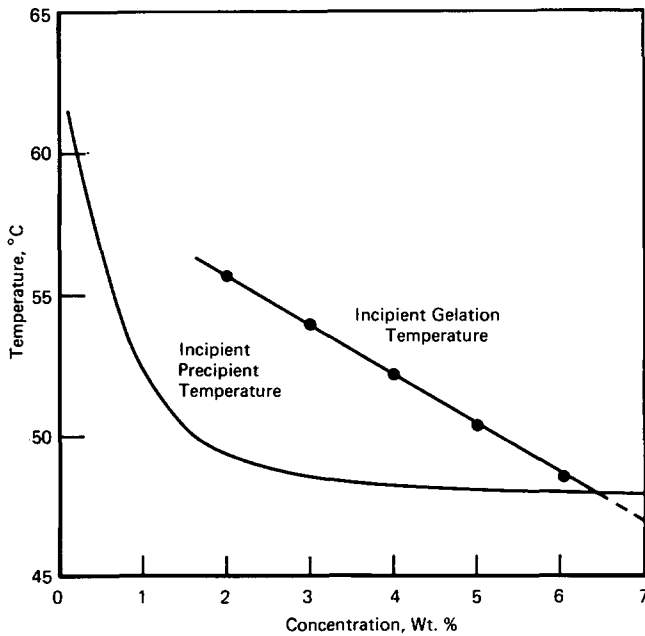


Fig. 4. Temperature of sol-gel transformation for aqueous solutions of Methocel F50 as function of concentration.

in about 3 hr, after which there was no change in strength. In general, when the gel is allowed to stand at that temperature for an extended period of time (more than a day), an observable syneresis of the gel takes place because of shrinkage

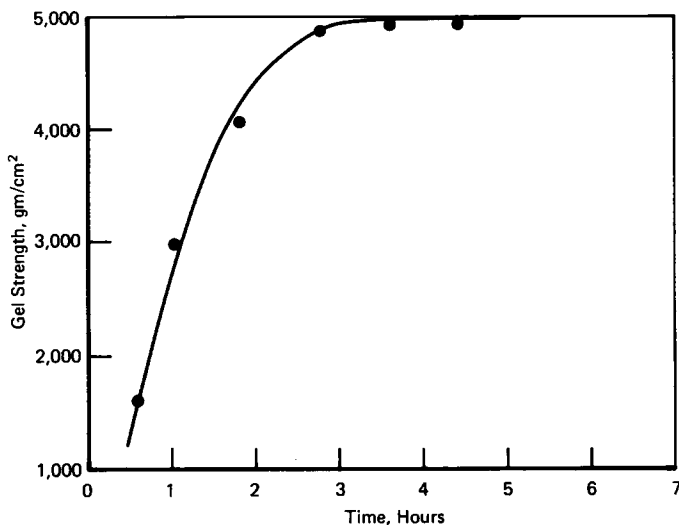


Fig. 5. Rate of gel strength development for 2% solution of Methocel A4C upon heating at 65°C.

of the three-dimensional structure. The syneresis is highly affected by temperature; and the higher the temperature and electrolyte concentration, the faster is the syneresis. So care was taken to measure gel strength before any syneresis was observed.

Gel characteristics of methylcellulose are known to vary considerably with type and degree of substitution, concentration, molecular weight, additives, etc. Depending on the type and total degree of substitution, both the gelation temperature and gel strength can vary considerably. In the following paragraphs some of these properties are discussed.

Effect of Molecular Weight

Table III shows the effect of molecular weight on the incipient gelation temperature, incipient precipitation temperature and the cloud point for 2% aqueous solutions of methylcellulose. It is interesting to note that within the molecular weight range studied there is no noticeable difference in the IGT, IPT, or CP. This is contrary to the classical polymer chemistry where one expects an increase

TABLE III
Gelation and Precipitation Temperatures of Methocel A Products as a Function of Molecular Weight

Molecular weight M_w	2% Solution viscosity at 20°C, cP	Gel point, °C	IPT, °C	CP, °C
~ 20,000	5.1	49.0	—	—
~ 38,000	15	—	43	63
~ 50,000	27.0	48.5	47	62
~ 90,000	98	48.5	—	—
~140,000	400	49.0	47	62
~400,000	4000	48.5	48	61

in precipitation temperature with decrease in molecular weight. Assuming that the concentrations of the solutions were too high to show true fractionation of the molecules, precipitation temperatures of Methocel A25 and Methocel A4M were measured at a concentration of 0.25%. The IPT and CP for Methocel A25 were 62°C and 85°C, respectively, while those for Methocel A4M were 63°C and >100°C, respectively. This is again the reverse of what one might expect for a normal polymer. This anomalous behavior of methylcellulose solution can however be explained if one takes into account the molecular weight distribution and the state of aggregation of the polymer in solution.

All the methylcellulose samples have a wide molecular weight distribution where the ratio of weight- to number-average molecular weight may vary from 3 to as high as 10 depending on the type of pulp used and the processing conditions. The incipient gelation temperature and the incipient precipitation temperature actually reflect the influence of the high molecular weight fraction of the sample which precipitates out first. All the samples of varying average molecular weight contain similar high molecular weight fractions, although of varying amounts. This is the reason why all these samples are exhibiting similar gelation and precipitation temperatures.

On the other hand, the anomaly observed at low concentration by low molecular weight methylcellulose (Methocel A25) exhibiting lower cloud point than the high molecular weight sample (Methocel A4M) can only be explained by the presence of aggregates in solution. Neely¹² and Kuhn et al.¹³ observed that aqueous methylcellulose solution even at room temperature contains aggregates. Kuhn et al.¹³ suggested that the low molecular weight methylcellulose, because of its rod-like structure, can aggregate more via parallel arrangement along the chain than the high molecular weight polymer, having a random coil configuration. This can result in the observed lowering of cloud point when the molecular weight is decreased.

In an attempt to eliminate the aggregates in solution by cooling, two methylcellulose solutions of 2% concentration and having viscosity of 26000 cP and 10 cP, respectively, at 20°C were stored in a refrigerator at 4°C for a week just prior to precipitation temperature measurement. While the incipient precipitation temperature remained the same at 43.5°C for these two samples, the cloud points of 26000-cP and 10-cP methylcellulose samples were 56.5 and 59°C, respectively. This confirms the aggregation theory and indicates that the aggregates can be dispersed, if not completely, by cooling the solutions.

Gel strength of methylcellulose solution, on the other hand, is dependent on molecular weight, as shown in Figure 6. The gel strength increases with increasing molecular weight and gradually levels off at or above an average molecular weight of 140,000 (Methocel A4C). Compared to these results, a linear relationship is found between the logarithm of viscosity and the logarithm of molecular weight. While this independence of gel strength above a certain molecular weight is difficult to explain from a mechanistic point of view, these results are similar to those observed with gelatin.¹⁴ These results are also analogous to the relationship between the strength of native cellulose fibers and their DP (degree of polymerization),¹⁵ in which the strength of acid-treated cotton increases with DP and then finally levels off at a DP between 700 and 1000. Surprisingly, the molecular weight of 140,000 for methylcellulose also corresponds to a DP of 760.

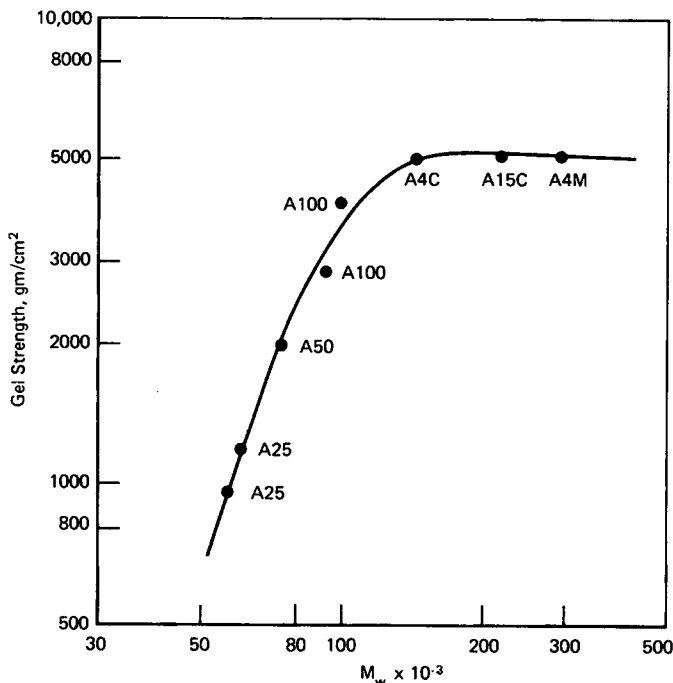


Fig. 6. Gel strength of 2% aqueous methylcellulose gels after 4 hr at 65°C as function of molecular weight. Numbers designate Methocel A samples of different viscosity grades.

Effect of Substitution

Table IV shows the precipitation temperatures of different hydroxypropyl methylcellulose samples having DS and MS as shown in Table I, and of different molecular weight. Here again no significant difference in IPT is observed, indicating the influence of molecular weight distribution. On the other hand, cloud points show a normal decrease with increase in molecular weight, indicating that the aggregation phenomenon in these samples is not as predominant as was observed with methylcellulose samples. Effect of total degree of substitution and the hydroxypropyl substitution is also observed as the cloud point, for the similar viscosity-grade products, decreases in the order $K > F > E$ products, and the values are larger than observed with methylcellulose samples.

TABLE IV
Precipitation Temperatures of 2% Aqueous Solutions of Different Methocel Brand Cellulose Ethers Having Different Molecular Weight and Substitution

Methocel brand	IPT, °C	CP, °C
E15	61	67
E50	61	64
E4M	58	61
F50	61	69
F4M	60	65
K35	61	79
K100	60	76
K4M	61	70

It is quite well known that the type of substitution is important in determining the gelation properties of cellulose ethers. For example, carboxymethyl cellulose, sulfoethyl cellulose, and hydroxyethyl cellulose are all nongelling types. Hydroxypropyl cellulose does not show gelation but does precipitate out of solution on heating. Hydroxyethyl methylcellulose, containing a high degree of methoxyl and a low degree of hydroxyethyl substitution, is known to exhibit gelation, but the polymer containing a high degree of hydroxyethyl and a low degree of methoxyl substitution does not gel. Apparently, while it is the methoxyl substitution in hydroxypropyl methylcellulose which is responsible for its gelation, the hydroxypropyl substitution is also responsible for altering the gelation characteristics significantly.

Figure 7 shows the incipient gelation temperature-versus-concentration relationship for three different Methocel brand products. While the relationship is linear for all, their slopes are quite different. All these products have a similar methoxyl degree of substitution (1.7–1.9), while Methocel A, F, and E have hydroxypropyl molar substitution of 0, 0.13, and 0.22, respectively. These data indicate that the differences in IGT at low concentrations are not very significant. But as the concentration increases, IGT diverge. Thus, methylcellulose (Methocel A) gels at room temperature when the concentration reaches about 12%, while the hydroxypropyl methylcellulose gels at room temperature only when the concentrations are significantly higher.

If one maintains the hydroxypropyl substitution constant, say, at about MS = 0.22 and lowers the methyl substitution, the gel point increases gradually due to lower total degree of substitution and hence lower hydrophobe–hydrophobe interaction. For example, Methocel K (methoxyl D.S. = 1.1–1.4, and hydroxypropyl M.S. = 0.1–0.3) gels at 70–90°C. It has also been observed that lowering the methoxyl substitution produces gels that are mushy in character with yield value so low that data could not be generated using the cone penetrometer.

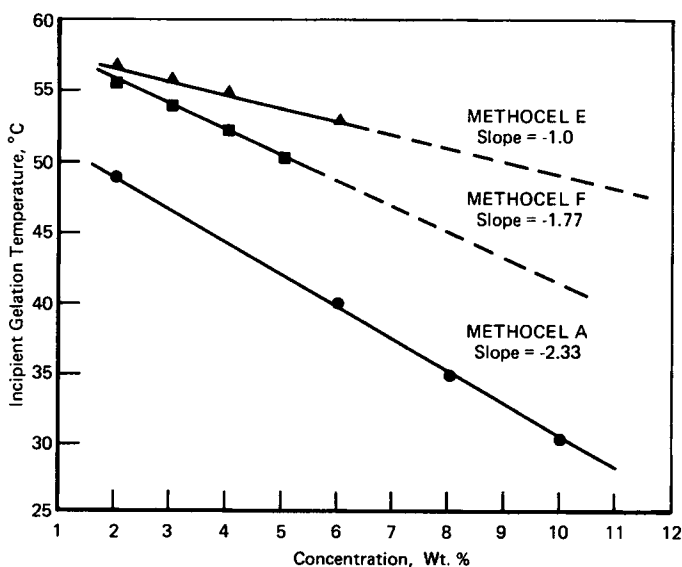


Fig. 7. Incipient gelation temperature of different Methocel products as function of concentration.

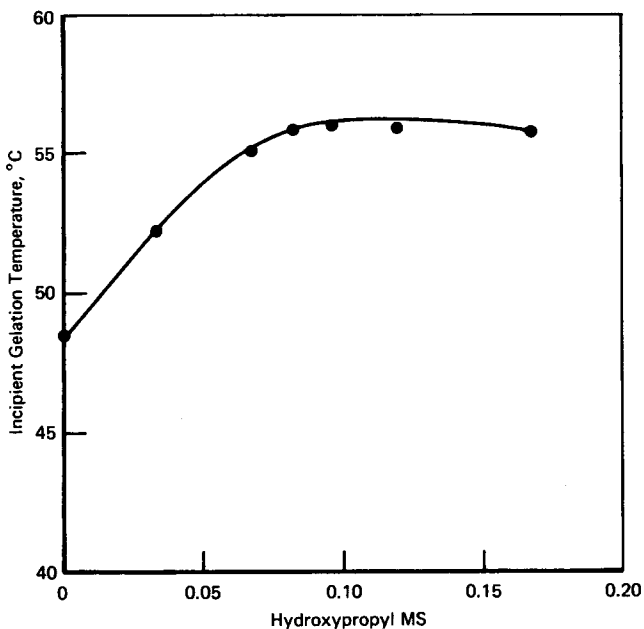


Fig. 8. Incipient gelation temperatures of hydroxypropyl methylcellulose as function of hydroxypropyl molar substitution. Methoxyl DS = 1.63–1.88; 2% solution viscosity = 400–8000 cP.

Figure 8 shows the effect of hydroxypropyl substitution (MS) on IGT for 2% solutions of hydroxypropyl methylcellulose. In this experiment methoxyl DS was kept at 1.63–1.88, and the 2% solution viscosity of the samples at 20°C were between 400 and 8000 cps. Since IGT is not dependent on molecular weight at these viscosity ranges, the data actually show the effect of hydroxypropyl MS. It is seen from Figure 8 that as the hydroxypropyl MS is increased, the IGT increases slightly, reaches a maximum at about 0.1 MS, and then decreases very slightly with increasing MS. Overall, there is not a drastic change in IGT between 0 and 0.2 MS.

In contrast, the effect of hydroxypropyl MS on gel strength is significant, as shown in Figure 9. Straight methylcellulose with a DS of 1.63–1.88 has a very high gel strength. As the hydroxypropyl substitution increases, the gel strength decreases drastically until at MS = 0.15 and above the gels become mushy.

These results are a little difficult to explain from the point of view of mechanism. Both the methoxyl and hydroxypropyl substitutions render the cellulose hydrophobic. The higher gelation temperature and lower gel strength of methylcellulose upon increasing hydroxypropyl substitution can therefore be due only to steric reasons. On the other hand, keeping the hydroxypropyl substitution constant while decreasing the methoxyl substitution renders the polymer hydrophilic because of decreased total degree of substitution. This also results in increased gelation temperature and decreased gel strength. Thus, by varying the type and DS of the substituent groups, various temperature behaviors of hydroxypropyl methylcellulose can be achieved.

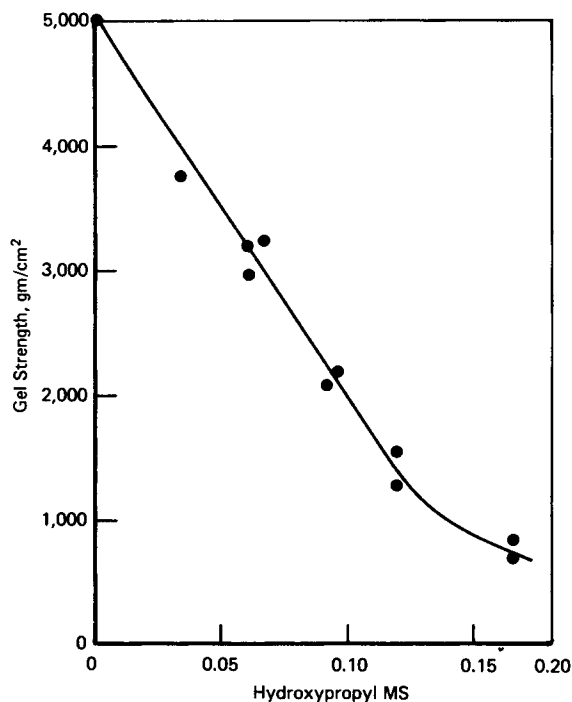


Fig. 9. Gel strength of 2% aqueous HPMC gels after 4 hr at 65°C as function of hydroxypropyl molar substitution. Methoxyl DS = 1.63–1.88; 2% solution viscosity = 400–8000 cP.

Effect of Additives

Gelation properties of methylcellulose can be substantially altered by the use of additives.^{16,17} Most electrolytes, as well as sucrose, glycerol, etc., depress the gel point owing to their greater affinity for water, thus dehydrating the polymer. A few compounds, such as ethanol and propylene glycol, act to elevate the gel point. Although these compounds have greater affinity for water than the cellulose polymers, they act as solubilizers due to their solvent power (or cohesive energy density) and increase the gelation temperature.

The precipitation temperature of a methylcellulose solution is also known to increase in the presence of urea.¹⁸ Water is normally hydrogen bonded to the polymer. Urea molecules act on the hydrogen bond and attach themselves to the polymer by displacing water molecules. Dissociation of the urea molecules takes place at a considerably higher temperature than the dehydration temperature.

In this paper the effect of sodium chloride on the gel point and gel strength is described in more detail. Figure 10 illustrates the effect of sodium chloride on the gel point, incipient precipitation temperature, and gel strength (after heating at 65°C for 3 hr) for a 2% methylcellulose solution, indicating that sodium chloride considerably lowers the gelation and precipitation temperature of the solutions. On the other hand, there is a significant increase in gel strength on increasing salt concentration.

This reduced solubility behavior of organic nonelectrolyte in water by the addition of neutral salts is quite well known. This "salting out" was once thought of as due to the competition between the polymer and the electrolyte for the water

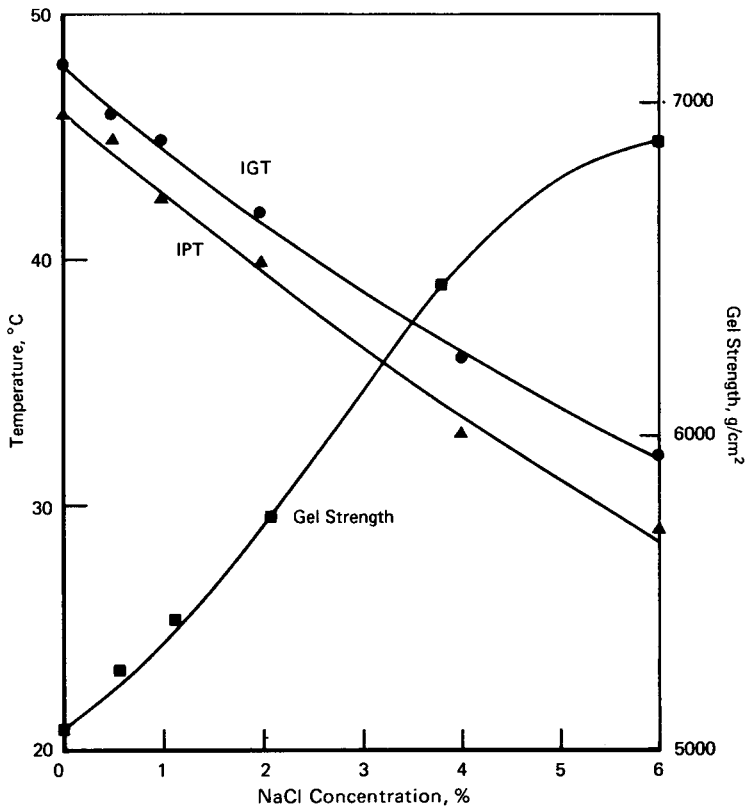


Fig. 10. Incipient gelation temperature (IGT), incipient precipitation temperature, and gel strength of 2% methylcellulose (Methocel A15C) solution as function of NaCl concentration.

molecules. However, the phenomenon is more complex, since the relative "salting out efficiency" of different salts varies widely with the nonelectrolyte polymer.¹⁹ Solvent power of electrolyte solution depends strongly on both cations and anions, increasing with the polarizability of the anion. Stanton et al.²⁰ studied a series of electrolytes and tabulated them in terms of solvent power of these aqueous salt solutions for nonelectrolyte polymers. Utilizing their table, the gelation properties of methylcellulose solution can also be approximately predicted as a function of electrolyte types. Any additive that increases the solvent power of water for the polymer would elevate the gelation temperature, and vice versa.

In conclusion, we now have a better understanding of the gelation properties of methyl and hydroxypropyl methylcellulose. However, we are still a long way from a complete understanding of these very interesting properties and their correlation with varied end use applications. Such studies will be continued and reported at appropriate intervals.

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